

Self-assembled periodic sub-wavelength structures by femtosecond laser direct writing

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Abstract: Self-assembled, sub-wavelength periodic structures are induced in fused silica by a tightly focused, linearly polarized, femtosecond laser beam. Two different types of periodic structures, the main one with period (Λ_E) in the direction of the laser beam polarization and the second with period (Λ_k) in the direction of the light propagation, are identified from the cross-sectional images of the modified regions using scanning electron microscopy. We demonstrate the spatial coherence of these nanogratings in the plane perpendicular to the beam propagation direction. The range of effective pulse energy which could produce nanogratings narrows when the pulse repetition rate of writing laser increases. The period Λ_E is proportional to the wavelength of the writing laser and period Λ_k in the head of the modified region remains approximately the wavelength of light in fused silica.

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OCIS codes: (320.2250) Femtosecond phenomena; (260.1440) Birefringence.

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1. Introduction

Femtosecond (fs) lasers, with their ability to induce a refractive index modification inside bulk materials, have proved to be a successful tool for fabricating three-dimensional photonic devices, optical data storage and bio-photonic components in a variety of optically transparent materials [1-5]. Moreover, since the physical mechanism inducing a refractive index change in the irradiated material is based on nonlinear absorption, complex structures can be directly fabricated in large band gap materials such as pure silica, traditionally challenging for direct laser processing. Furthermore, depending on the laser intensity, different features with either positive or negative index changes or voids can be realized in the focal volume of fused silica, thus enabling numerous applications such as waveguides [1], diffractive optics [6] and micro-fluidic channels [7].

From our previous experimental work, it has been shown that structures written in fused silica by fs laser pulses above a certain intensity threshold exhibit anisotropic reflection [8]. At that time, we speculated that its appearance, only in the direction of the polarization of the writing laser beam, could be explained by assuming that a sub-wavelength period grating is formed within the irradiated focal volume. This explanation was further strengthened by the consideration that the orientation of the suspected nanogratings is identical to that implied by the empirical observation of anisotropic light scattering in our previous work [9]. The first experimental evidence of self-assembled nanostructures created within irradiated regions in fused silica was presented in [10]. This type of periodic structure, whose period is smaller than the laser's wavelength, explained the concurrent appearance of anisotropic reflection [8], a permanent birefringent microlayer [11], form birefringence, and an average negative index change [12]. Embedded micro-reflectors, retardation plates, and micro-fluidic channels based on these nanostructures have been reported [6,8,13,14]; however their optimization requires a systematic characterization of the properties of the periodic structures versus various fabrication parameters. Recently, changes in the nanogratings due to variation of the pulse duration have been qualitatively investigated [15]. The extraordinary stability of these nanogratings has also been reported [16].

Here we report on the investigation of nanogratings in the light propagation direction and in the perpendicular plane. In addition, we complete the quantitative analysis of their period under various laser parameters.

2. Experiments

Two different types of laser sources and three 1 mm thick fused silica (sample A, B, C) have been used in experiments. The first laser source was a regeneratively amplified mode-locked Ti:Sapphire laser system (Coherent RegA) emitting a train of pulses with pulse duration (τ_p)

varied from 150 to 250 fs, at repetition rate (R_{ep}) of 250 kHz and at wavelength (λ) tunable from 750 nm to 850 nm. Prior to focusing, the laser beam was linearly polarized along the x- or y-axis (shown in Fig. 1(a)) using a half-wave plate, and the pulse energy (E_p) was varied using a variable neutral density filter from 50 nJ to 1 μ J. The laser beam was focused via a 50 $\times$ objective (NA=0.55) $\sim$ 200 μ m below the surface of sample (sample A and sample B). Straight lines were written by translating the sample along the y-axis, perpendicular to the propagation direction of the laser beam (z-axis), as shown in Fig. 1(a). A Fresnel zone plate B (FZP_B), described in [6], was also chosen for the study of nanogratings in the plane perpendicular to the propagation of the writing laser. It consisted of a series of concentric rings written with similar conditions to sample A.

The second laser source was a variable repetition rate fiber laser system (IMRA America FCPA μ Jewel D-400). Using a similar experimental setup, the sample C was irradiated under various repetition rates (200 kHz, 500 kHz and 1 MHz) and at two different wavelengths (1045 nm and 522 nm). The pulse duration was 400 fs and the beam was focused via a 30 $\times$ single element aspheric lens (1045-nm processing) and a 40 $\times$ microscope objective (522-nm processing), both at an operating NA of $\sim$ 0.32.

After laser irradiation, the sample A was analyzed using scanning electron microscope (SEM) (JSM6500) after chemical etching. The samples B, C and Fresnel zone plate B were imaged using backscattering SEM without etching.

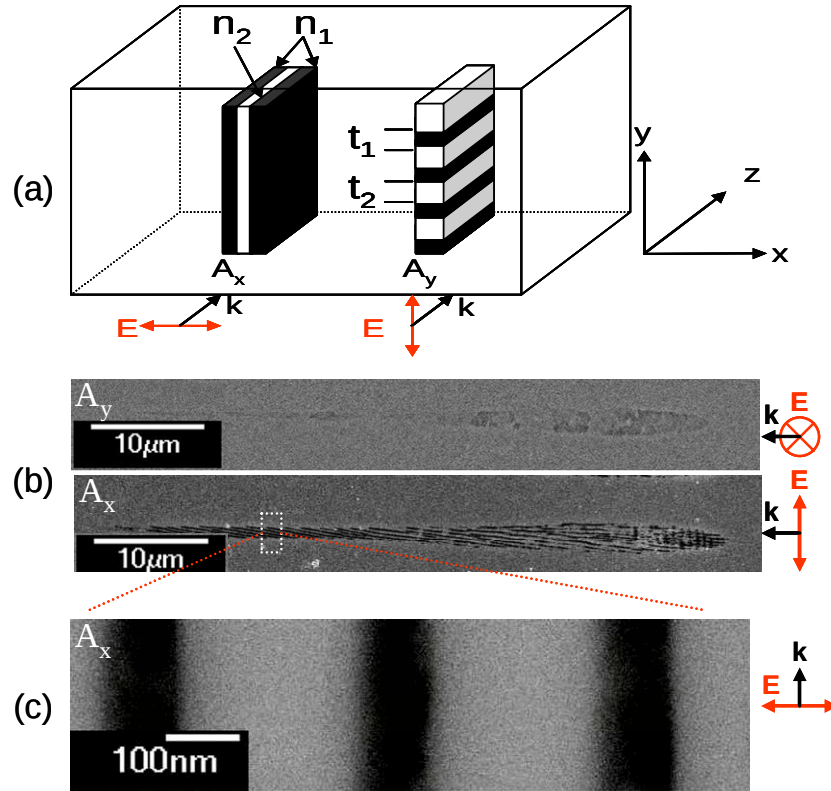


Fig. 1. Modified regions in sample A. E: electric field of the writing laser, k: wave vector of the writing laser beam, line A_x and line A_y : $E_p = 0.48 \mu\text{J}$, $V = 200 \mu\text{m/s}$, $\lambda = 850 \text{ nm}$. (a): Schematic of the sample A showing how the self-assembled periodic structures are expected to be arranged. n_1 and n_2 : local refractive indices of the plates of thickness t_1 and t_2 , respectively. (b): SEM images of A_x and A_y in the xz plane. (c): Detail of the sub-wavelength periodic structure formed in the cropped region of A_x .

3. Results

As depicted in Fig. 1(a), only the line A_x written with the polarization along x-axis (Fig. 1(b)), in the plane of observation (xz) reveals the periodic modulation of the index change (for these structures, xz is the plane perpendicular to the plates forming the nanogratings). On the contrary, the corresponding line A_y , written with the same conditions, but with the polarization along y-axis (Fig. 1(b)) shows a homogeneous profile (for these structures, xz is the plane parallel to the plates forming the nanogratings). Figure 1(c) is a close-up of part of Fig. 1(b) revealing that the filling factor f is ~ 0.3 , twice as high as the results reported in [8] (the filling factor is defined as $f = t_1 / \Lambda$, with t_1 the thickness of the zones of refractive index n_1 in Fig. 1(a), and Λ the period of the nanograting). In addition, the SEM image of the etched sample reveals the surface relief created in the etching process due to the density variations in the gratings. Figure 1(c) demonstrates an abrupt change of density between dark area with low density of the material (i.e. the zones of refractive index n_1 in Fig. 1(a)), and the surrounding regions (i.e. the zones of refractive index n_2 in Fig. 1(a)), suggesting that the mechanism leading to the modulation of the refractive index is highly nonlinear. Furthermore, the mechanism of the structural changes responsible for the nanograting formation involves the modulation of the electron concentration, negatively charged oxygen ions can be repelled from the dark area with high electron concentration [10].

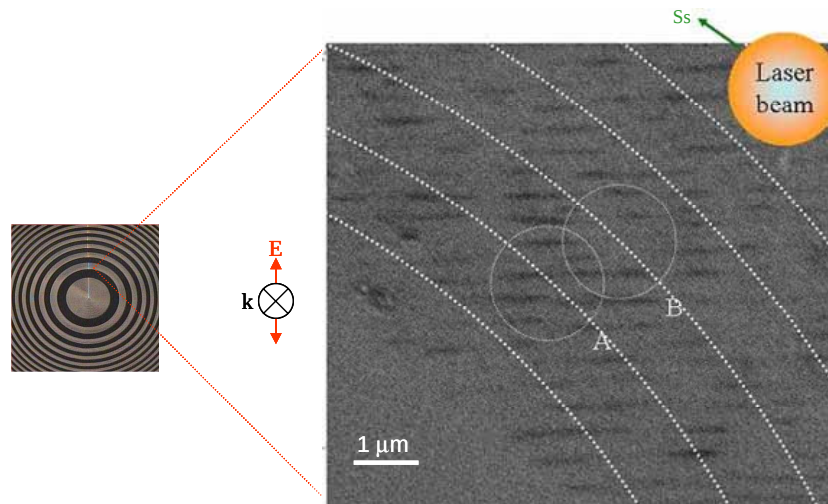


Fig. 2. SEM image of the nanogratings in the Fresnel zone plate B. E: electric field of the writing laser, k: wave vector of the writing laser beam, Ss: movement direction of the translation stage.

Evidence of spatial coherent of the nanogratings in the plane perpendicular to the propagation of the writing laser is provided by backscattering electrons SEM image of Fresnel zone plate B, which is sensitive to the atomic weight of the elements or the density of the material constituting the observation surface. As expected, the grating ruling is perpendicular to the direction of the polarization of the writing laser beam. In addition, the image clearly shows that the self-assembled nanostructures formed in adjacent lines are aligned. As described in [6], the Fresnel rings were realized by writing circles with a space between them smaller than the writing resolution; hence the processed zones are formed by a series of partially overlapped directly written circles (Fig. 2). Given the geometry of the lens and the speed of the translation stage, the points in B in Fig. 2 were written ~ 2 s after the points in A. Despite the long time elapsing between the formations of the periodic structures in the two regions, the nanograting is continuous across the two written lines. This evidence suggests that the structure imprinted in a line provides an initial “seeding” condition during

the formation of the self-organized periodic assembly in the adjacent line. This is a very important property as it demonstrates the ability of realizing periodic nanostructures over a larger area, rather than being limited to the size of the focused spot.

Figure 3 presents SEM images of the modified region in the xz plane induced by three different writing laser wavelengths in sample B and sample C. The periodic structures can be observed in the head of the modified regions. Besides the main period Λ_E which is in the direction of the polarization of the laser beam (E), a second period Λ_k is identified perpendicular to Λ_E , and hence parallel the direction of propagation of laser beam (k), as shown in Fig. 3. Furthermore, Λ_k is 710 ± 10 nm at 1045 nm (Fig. 3(a)), 550 ± 10 nm at 800 nm (Fig. 3(b)), and 360 ± 10 nm at 522 nm (Fig. 3(c)) in the head of the modified region, which is very close to the ratio λ/n in fused silica.

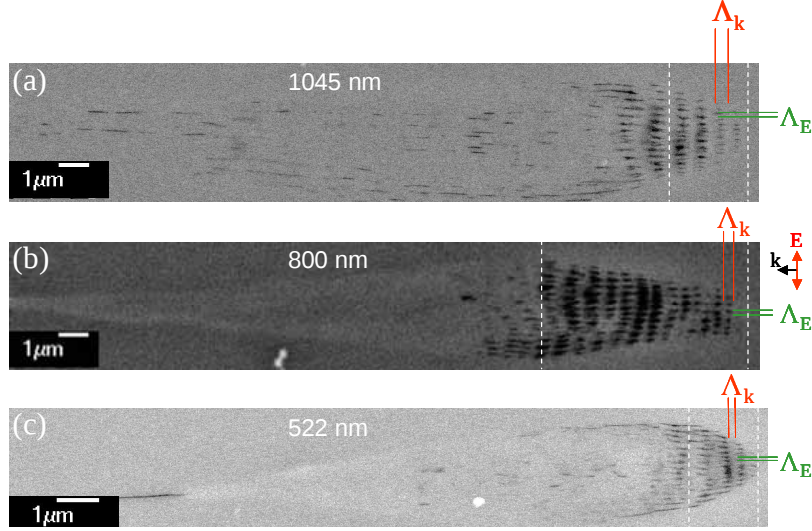


Fig. 3. SEM images of nanogratings formed by three different central wavelengths of the irradiated laser pulses in sample B and sample C, the region between two dotted lines is used for period calculation, E: electric field of the writing laser, k: wave vector of the writing laser beam. (a): $\tau_p = 520$ fs, $E_p = 0.9$ μ J, $V = 200$ μ m/s, $R_{ep} = 500$ kHz. (b): $\tau_p = 150$ fs, $E_p = 0.5$ μ J, $V = 100$ μ m/s, $R_{ep} = 250$ kHz. (c): $\tau_p = 490$ fs, $E_p = 0.15$ μ J, $V = 200$ μ m/s, $R_{ep} = 200$ kHz.

In order to study the nanogratings under various repetition rate regimes, lines structures induced in sample C were inspected using the Nomarski-DIC microscope. It shows that, when the repetition rate of the laser pulses reaches 500 kHz and E_p is over ~ 1 μ J (Fig. 4(a)), or when the repetition rate reaches 1 MHz and E_p is over ~ 0.6 μ J (Fig. 4(b)), no birefringence or nanogratings could be observed in the irradiated regions of the sample. This could be explained by the occurrence of an accumulation effect within the focal volume when the pulse repetition rate is greater than ~ 500 kHz [17,18]. This indicates that the range of the pulse energy which could produce self-assembled nanogratings narrows when the pulse repetition rate of the laser increases.

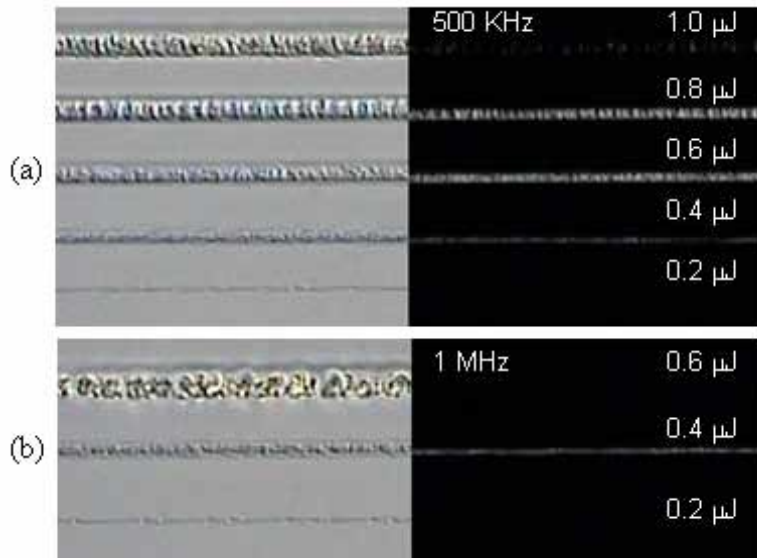


Fig. 4. Microscope images of the irradiated regions in the xy plane in sample C, image on the left taken by back-illumination without polarizers, image on the right taken by back-illumination with cross polarizers of the same irradiated area. (a): $R_{ep} = 500$ kHz, $V = 500$ $\mu\text{m/s}$. (b): $R_{ep} = 1$ MHz, $V = 1000$ $\mu\text{m/s}$.

In order to quantitatively analyze the period Λ_E and Λ_k , an algorithm based on the correlation coefficients between different points in the SEM image versus their distance was developed. By calculating the average value of the periods from the first maximum of the correlation functions, periods along the x and z direction are obtained, shown in Fig. 5.

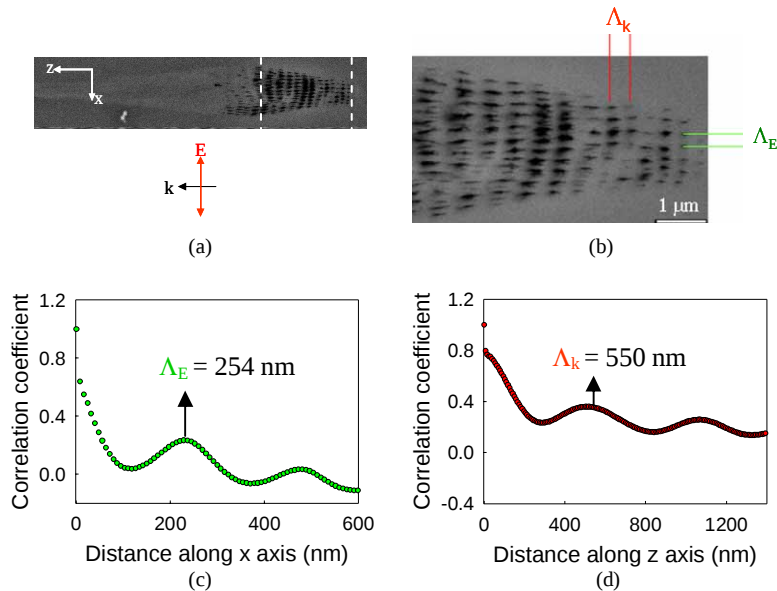


Fig. 5. (a): SEM image of a written line in the xz plane, E: electric field of the writing laser, k: wave vector of the writing laser beam ($\lambda = 800$ nm, $\tau_p = 150$ fs, $E_p = 0.5$ μJ , $V = 100$ $\mu\text{m/s}$, $R_{ep} = 250$ kHz). (b): The region between two dotted lines is used for calculation. Corresponding normalized correlation functions calculated along the x (c) and z (d) axis.

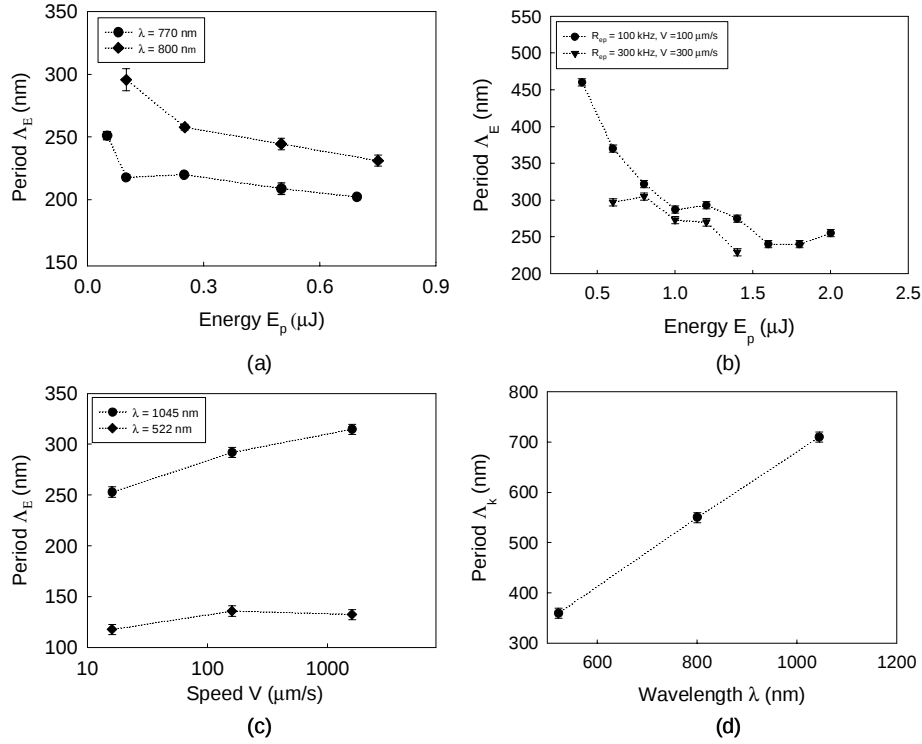


Fig. 6. (a): Period Λ_E versus E_p ($\tau_p = 150$ fs, $R_{rep} = 250$ kHz, $V = 100$ μ m/s). (b): Period Λ_E versus E_p ($\tau_p = 480$ fs, $\lambda = 1045$ nm). (c): Period Λ_E versus speed V at two different wavelengths ($\tau_p = 400$ fs, $R_{rep} = 200$ kHz, $E_p = 0.5$ μ J). (d): Period Λ_k versus three different wavelengths.

Our analysis of numerous SEM images reveals that the period Λ_E and Λ_k does not change with the scanning speed from 50 μ m/s to 500 μ m/s. Moreover, Λ_E decreases with increasing E_p within our study range (Fig. 6(a-b)). In addition, the wavelength dependence illustrated in Fig. 6(a) suggests that shorter wavelengths yield significantly smaller structures; this is also confirmed in Fig. 6(c), which demonstrates that the period Λ_E is proportional to the wavelength of the writing laser. Furthermore, Λ_k in the head of the laser-modified region does not change with the pulse energy or speed and is confirmed to be approximately equal to the laser wavelength (Fig. 6(d)).

Our previous experimental results showed that over a certain intensity threshold, these femtosecond laser induced periodic structures behave as an uniaxial form-birefringence material [12]. This is also confirmed using the FCPA μ Jewel D-400 at wavelengths of 1045 nm and 522 nm. Figure 7 shows a replica of IMRA's icon that was made by raster-scanning closely spaced lines (10- μ m spacing).

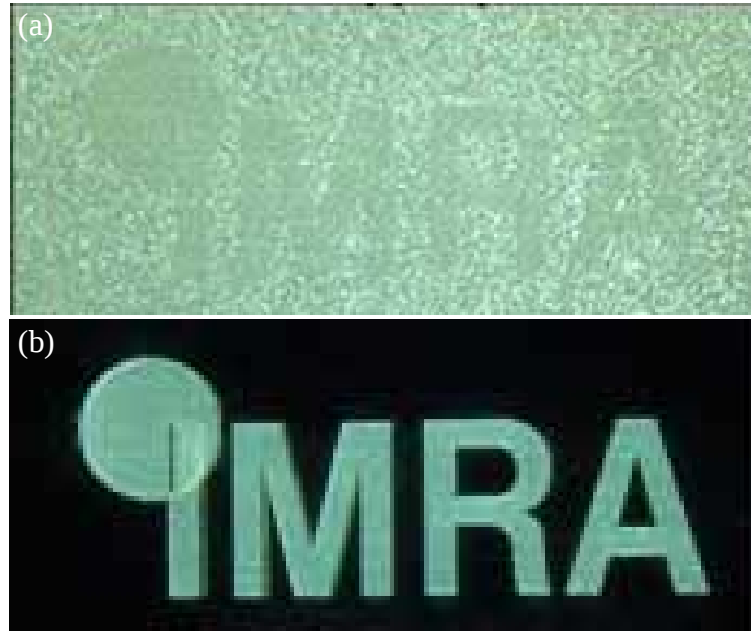


Fig. 7. Raster scanned replica of IMRA icon, 10- μm line spacing, 500-kHz repetition rate, 1045-nm wavelength, 1.5- μJ pulse energy, ~12-hour process time, ~10-mm icon width. (a) without cross-polarizers, (b) with cross-polarizers.

4. Conclusions

In conclusion, two different types of periodic structures have been identified from cross-sectional SEM images of femtosecond pulse-modified regions in fused silica. The range of pulse energy which could produce nanogratings narrows when the pulse repetition rate of the writing laser increases. The period Λ_E is proportional to the wavelength of the writing laser and the period Λ_k in the head of the modified region remains $\sim \lambda/n$ in fused silica. The spatial coherence of these nanogratings in the plane perpendicular to the beam propagation direction has also been demonstrated, which make them extremely attractive for new applications such as embedded micro-reflectors or photonic band-gap devices.